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X-RAY STUDY OF VEGETABLE FIBRES

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(Plates VIII A and VIII B)

ABSTRACT. Some vegetable fibres have been studied by means of X-rays and it is found that the orientation of cellulose crystallites is not same in all cases. In some fibres the major axis of the unit cell is almost parallel to the fibre axis and in some the major axis is at some spiral angle to the fibre axis while in an extreme case the cellulose crystallites are randomly distributed.

INTRODUCTION

Much progress has been made during the last few years regarding the knowledge of the structure of cellulose fibre from various sources of investigations—specially due to the correct interpretation of X-ray photograph, correlating the chemical information supplied by Haworth.

Cellulose is the main constituent of cotton, ramie, flax, jute, etc. and practically it builds up the frame-work of all vegetable tissues. It has been shown by various workers that the fundamental structures of cellulose derived from these various sources are identical. The elementary unit of cellulose is monoclinic with $a=8.35$A, $b=10.3$A and $c=7.9$A and $\beta=84^{\circ}$. The space group is C_2^2 with four glucose units in the unit cell. The cellulose fibre is a crystalline aggregate consisting of small crystallites separated by amorphous or intercrystalline areas. The crystal area is built up by repetition of the unit cell arrangement of glucose units in space.

X-ray investigation of jute fibre has been made by Banerjee and Roy, (1941) Sarkar, Rudra and Saha (1944) and Banerjee, Basak and Sen (1945). Some vegetable fibres were also studied by Bose and Ahmed (1946). In the present investigation a number of vegetable fibres—Pandanus., Annanus sativus, Sesbania aculeas, Hibiscus esculantus, Calotropis jigantea, coma on the seed of Calotropis jigantea, Hibiscus mutabilis, Musa sapientum, Crotalania juncea, Agave Americana, Coir A which had been isolated from green-cocoanut and coir B which had been isolated from dried cocoanut—have been studied by X-rays.

The fibres of Hibiscus esculantus and Crotalania juncea were, however, previously studied by Bose and Ahmed and the Coir B has been studied by Astbury. There are, however, considerable difference between the X-ray photographs of cellulose derived from different sources regarding the

diffuseness and directions of extensions of the spots indicating differences in the internal order among the ultimate fibres and extents over which regularity of arrangement occurs.

A narrow pencil of X-rays from a Hadding-Seigbahn type of X-ray tube with Cu anti-cathode run at a voltage of 40.KV and a tube current of 5-7 milliamps was incident on a thin bundle of fibre (about 1 mm. diameter) normal to the length of the fibre. The diffracted X-rays were received on a cylindrical film of radius 5 cm. Exposures of about 8 hours were given. The photographs obtained are reproduced in the Plates VIII A and VIII B.

The angular co-ordinates θ and μ of the spots were obtained in the usual way and the glancing angle was obtained from the following relation $\cos\theta \cos\mu = \cos\theta_K$. The identification of spots were made by comparing the experimental glancing angle with the calculated Bragg angles of various planes the index K being known from the layer line in which the spots occur.

Crotalaria Juncea

Indices	Intensity	Character
(101)	S	Sharp and well resolved
(002)	S	Sharp and distinct
(004)	m	Slightly extended along the radial direction
(311), (212)	S	Well resolved
(120), (021)	S	Sharp and distinct
(221), (122)	m	Slightly broad
(130)	m	Diffuse and broad
(032)	S	Sharp and extended along the radial direction
(232)	m	Diffuse

Calotropis gigantea

Indices	Intensity	Character
(101)	S	Sharp and long
$(10\bar{1})$	S	Sharp and long
$(002)_\beta$	m	Discrete line of uniform width
(002)	VS	Sharp and slightly bended in the form of an arc.
(004)	m	Elongated to a certain length.
(311) / (013)	S	Sharp and slightly bended.
(021)	S	Sharp and well resolved.
(221) / (122)	W	Diffuse.
(131)	m	Diffuse and broad.
(231) / (032)	S	Sharp and slightly bended along the arc of a circle
(331)	mw	Diffuse and elongated to a certain length along the radial direction.
(042) / (241)	m	Diffuse and elongated along the circumference of the diffuse ring

Sesbania aculeas

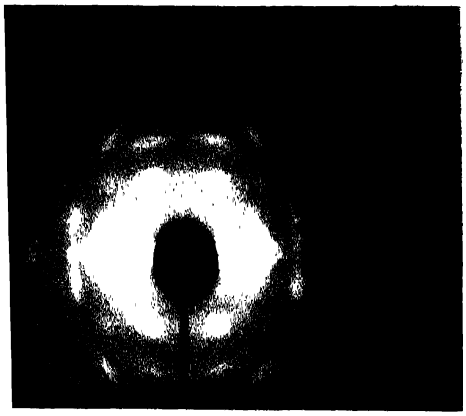
Indices	Intensity	Character
$(10\bar{1})$	S	Sharp and well resolved.
(002)	S	Distinct and bended along the arc of the circle on which the spots lie.
(004)	W	Very diffuse.
(013)	S	Bended to an arc.
(021)	S	Sharp and well resolved.
(130)	W	Broad and diffuse
(032) / (231)	S	Sharp and bended along the circumference of a circle on which the spots lie.

Hibiscus mutabilis

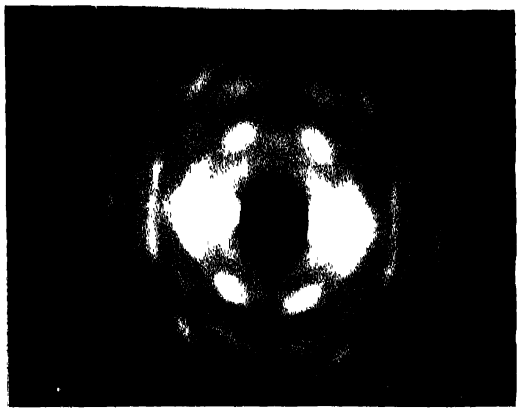
dices	Intensity	Character
(101)	S	Sharp and well resolved
(002)	S	Sharp and long
(004)	m	Very diffuse
(212) (311)	S	Drawn into a long arc
(021) (120)	S	Sharp and discrete
(130)	W	Diffuse and broad
(032)	S	Extended along the radial direction

Hibiscus esculantus

dices	Intensity	Character
(101)	S	Sharp and well-resolved
(002)	S	Sharp and bended to a long arc
(004)	W	Very diffuse
(212) (311)	S	Extended to a long arc
(120) (021)	S	Sharp and elongated along the radial direction
(130)	W	Diffuse and broad
(032)	S	Extended to a considerable length along the circumference of the circle on which the spots lie.



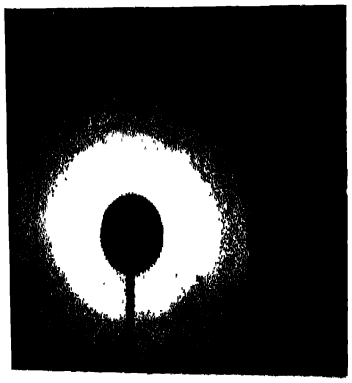
(1) Crotalaria juncea.



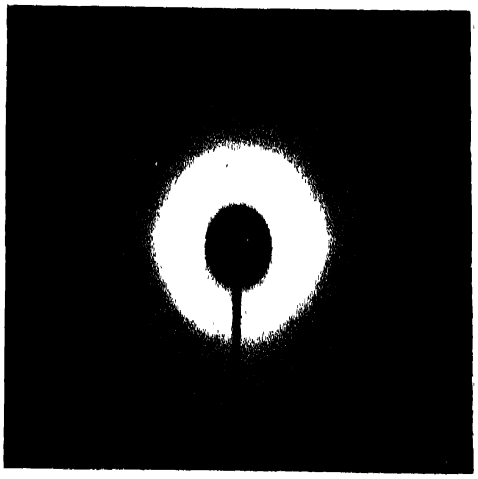
(2) Calotropis gigantea.

(3) Sesbania aculeas.

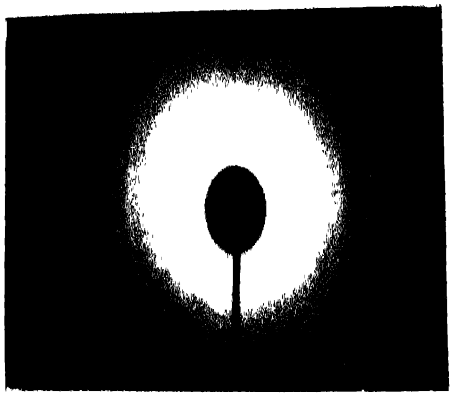
(4) Hibiscus mutabilis.



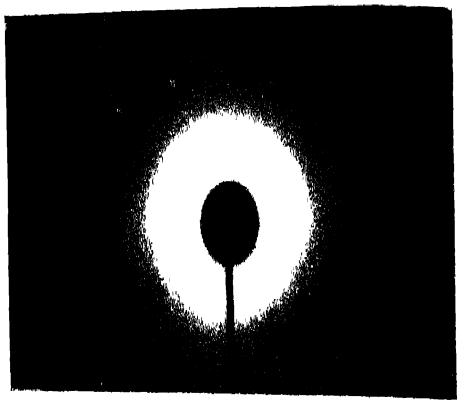
5) Hibiscus esculantus.



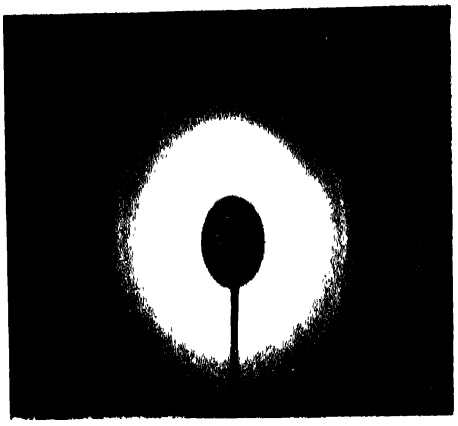
(6) Annonus salivus.



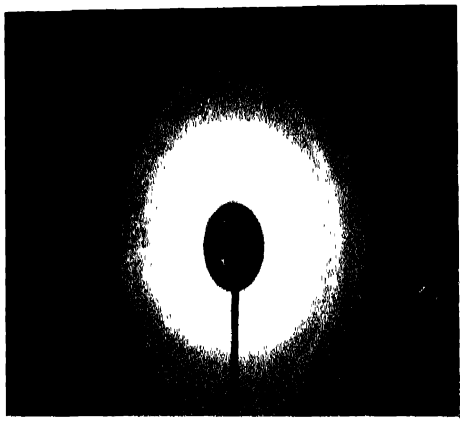
(7) Pandanus.



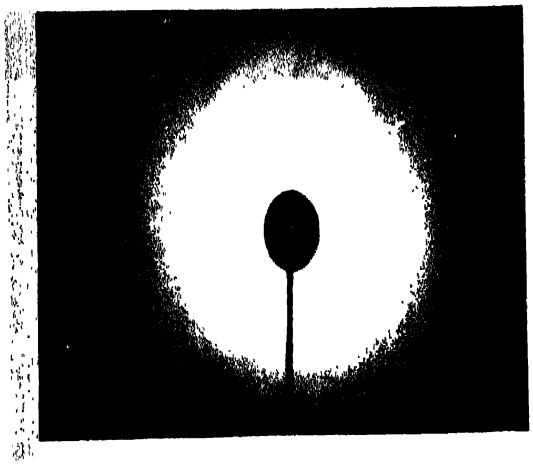
(8) Musa sapientum.



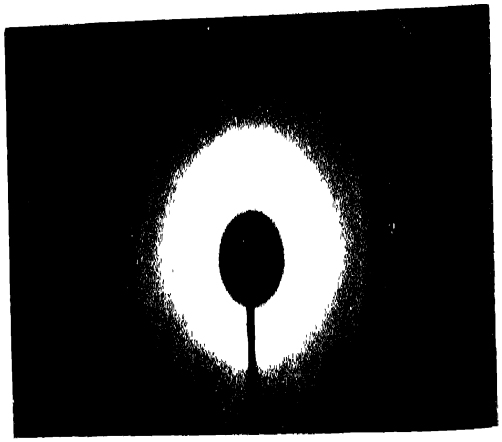
(9) Agave Americana.



(10) Coir-A.



(11) Coir-B.



(12) Coma on the seed of Calotropis gigantia.

Annanus Sativus

Indices	Intensity	Character
(101)	S	Sharp and well resolved
(002)	S	Sharp and bended along the radial direction
(212), (312)	m	Drawn into a longer arc
(021)	m	Not well resolved
(032), (231)	m	Drawn into a ring

Pandanus

Indices	Intensity	Character
(101)	S	Sharp and long
(002)	Vs	Sharp and bended to form a part of a ring
(103)	m	Marked intense portion of the continuous ring is taken as the position of the spot.
(004)	W	Diffuse

Musa sapientum

Indices	Intensity	Character
(101)	ms	Not well resolved
(002)	S	Sharp and extended to a considerable length along the radial direction.
(103)	m	Intense portion of the ring is taken as the position of the spot.

Agave Americana

Indices	Intensity	Character
(101)	S	Extended to a sharp arc
(002)	VS	Elongated into a sharp arc which is a part of a relatively less intense continuous circle.
(103)	m	Drawn into a complete ring of uniform intensity
(004)	W	Drawn into a continuous diffuse band

Coma on the seed of Calotropis gigantea

Indices	Intensity	Character
(101) (101)	m	Not distinct
(002)	S	Drawn into a complete ring of almost uniform intensity.
(103)	ms	Drawn into ring of uniform intensity
(004)	W	Diffuse band

DISCUSSION

It has been mentioned before that the cellulose is the chief constituent of all fibres. But the X-ray pictures of all fibres studied in this paper reveal that the degree of orientations of cellulose crystallites are not the same. The major axis of the unit cell of cellulose may be oriented either parallel to the fibre axis or at some spiral angle to the fibre axis or the crystallites may be randomly distributed. The example of each case mentioned above can be had in the X-ray photographs of various fibres in this paper.

The X-ray diffraction pattern of Crotalania juncea is similar to that of ramie or jute. It is just like the single rotation photograph. The spots are discrete and the resolution is perfect. The pattern of the picture as well as the shape and size of the spots indicate that the crystallinity extends over domains nearly of the same dimensions as those in ramie for which Mark and Meyer estimated an average dimension of about 50A thick but at least 600A long. The orientation of the '*b*' axis of crystallites also are approximately parallel to the fibre axis.

The X-ray diffraction pattern of Calotropis jigantea is very similar to that of above except that in this case there is a slight back-ground scattering

and the spots are rather broader and a little elongated. So the domains of crystallinity are slightly smaller than and the orientations of crystallites are rather inferior to those of Crotalania juncea.

The diffraction pattern of Sesbania aculeas and Hibiscus mutabilis have strong similarity to one another. The pattern and the positions of the spots are same as above but the shape and size of the spots are quite distinct from those of Crotalania juncea or Calotropis jigantea The spots retain then sharpness along the radial directions but extend along the directions of Debye-Scherrer rings so as to form arcs. This indicates that the orientation of crystallites of cellulose parallel to the fibre axis is partially deranged.

The pattern and the positions of the spots of Hibiscus esculantus are quite similar to those of Hibiscus mutabilis except that in this case the spots are drawn into still longer arcs. This indicates that the alingment of the crystallites of cellulose is considerably deranged from parallelism.

The X-ray diffraction pattern of Ammanus sativus is interesting. In this case the spots are completely drawn into arcs forming continuous Debye-Scherrer rings and the positions of the spots in the rings are indicated by the intense sharp portions in the rings. The spots on the equatorial line are quite sharp. All this indicates that there is a very wide disorientation of the crystallites of cellulose from parallelism to the fibre axis. The maxima of intensity on the rings, however, indicates that the tendency towards orientation still prevails.

There are some rings in the X-ray photographs of Pandanus and Musa sapientum and the spots on the equatorial layer line lie on the rings. Spots on the other layer lines are absent. This also indicates the disordered arrangement of the fibre molecule.

The diffraction pattern of Agave Americana produces some rings followed by diffuse bands. The first two rings are so marked with greater intensity on the equator that they give rise to two strong arcs which bend along the circumference of the respective ring. This picture is quite distinct from that of Calotropis jigantea or Hibiscus mutabilis where we get discrete spots in the diffraction pattern. This indicates that the cellulose crystallites are irregularly distributed in the fibre.

Coir A and Coir B give the same type of X-ray photograph. There are two strong arcs followed by diffuse rings but the ends of each arc are more intense than the middle portion of it. The distribution of intensity of the first arc of Coir A changes so abruptly at the ends that it gives rise to two spots It is known that in Coir B the crystallites of cellulose are at some spiral angle to the fibre axis. As the diffraction patterns of Coir-A and Coir-B are almost identical, so it can be legitimately concluded that in Coir A cellulose crystallites are at some spiral angle to the fibre axis.

In the X-ray pattern of Coma on the seed of Calotropis jigantea there are some rings with diffuse bands. This photograph is similar to the usual powder photograph. It indicates that the cellulose crystallites are randomly distri-

buted in this fibre. This picture is quite different from other fibres studied and is very similar to the young cotton fibre below 35-days where the evidence of cellulose orientation was absent.

The fibres studied in this paper have not been delignified and so there is a slight back ground scattering in every photograph.

Works are in progres to determine the size of cellulose miscelles as well as the nature and the degree of disorientation of cellulose crystallites to the fibre axis.

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